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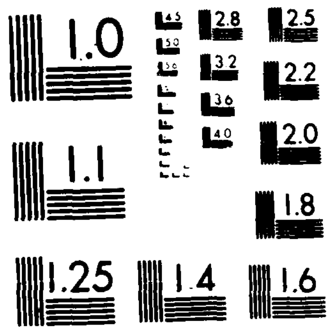

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The major progress in the second year of AFOSR research has been in the area of high-intensity laser beam interactions with single transparent micrometer-size droplets flowing in a linear stream. Motivated by our interest in in-situ and real-time diagnostics of the chemical species contained within droplets of sprays, we have continued the study of the nonlinear spectroscopy of single droplets. Having observed such nonlinear effects as lasing, stimulated Raman scattering, coherent anti-Stokes Raman scattering, and coherent Raman mixing when two input pump beams are present, we directed our research efforts toward exploring the limits of high-intensity irradiation of micrometer-size transparent droplets. Two effects ~~were~~ considered in detail: (1) the intensity-dependent index of refraction of the liquid, which can lead to self-focusing of the laser beam

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within the droplet and to broadening of the Raman radiation and elastic scattering due to phase modulation within the droplet; and (2) laser-induced dielectric breakdown, which can occur either in the gas surrounding the droplet or in the liquid within the droplet. The breakdown generates a shock wave and/or an optical detonation wave which causes material to stream from the droplet and form plumes. Progress in these areas will be described.

Significant success was achieved in developing an optical technique which has the potential of providing three-component velocity mapping in three dimensions. A time-coded five-pulse volume holography of silver-coated hollow spheres seeded in a cold flow has been recorded, and an automated pattern recognition technique (Hough transform) has been adapted to identify straight lines formed by the five exposures of the same silver-coated particles, which were sparsely seeded in a nozzle flow. Progress in this area will also be discussed.

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INVESTIGATION OF SUPERRADIANT LDV MARKERS
AND THREE-COMPONENT VELOCITY MAPPING

AFOSR GRANT NO. F49620-85-K-0002

January 1, 1986 - December 31, 1986

Richard K. Chang, Marshall B. Long, and Roman Kuc

Principal Investigators

Yale University
New Haven, Connecticut 06520

January 1987

RESEARCH OBJECTIVES

Following is a brief description of our three principal research objectives.

1. To investigate the feasibility of using bright lasing markers for flow diagnostics. The wavelength-shifted emission from the lasing markers can be optically isolated from the nonwavelength-shifted background caused by the turbulent medium and/or container walls. The laser emission from the droplets emerges in all directions and thus does not place a restriction on the observation angle as in the case of conventional markers which rely on the elastic scattering from the markers.
2. To investigate the possibility of using in-situ and real-time nonlinear optical spectroscopy [such as stimulated Raman scattering (SRS), coherent anti-Stokes Raman scattering (CARS), and coherent Raman mixing (CRM)] to provide molecular specific information within a single droplet. Because these nonlinear optical diagnostic techniques have successfully provided species-specific and temperature information in gaseous samples, it is desirable to extend similar nonlinear spectroscopic techniques to determine the chemical species contained in flowing single droplets, e.g., the droplets belonging to the spray of a two-phase combustor. Since increased input intensity leads to an improved signal-to-noise ratio in the spectra, the laser-induced breakdown limits of the liquid and the gas surrounding the droplet need to be understood.
3. To explore the possibility of combining time-coded multipulse laser volume holography and optical pattern recognition schemes to

determine the spatial displacement of many markers in a large sample during t, $t + \Delta t$, ..., $t + 5\Delta t$. Simultaneous determination of the three components of velocity from multiple points in a three-dimensional flow in combustion/turbulence diagnostics can be achieved by this approach.

RESEARCH STATUS

Our research results for the second year of the project can be summarized as follows:

1. Lasing Droplets

Color photographs of individual droplets undergoing lasing and SRS action have appeared in two journals [Science (publication #1) and in Optics News (publication #2)]. Such magnified photographs of droplets (radius $a \approx 35\ \mu m$) clearly reveal that the coherent radiation is confined just within the liquid-air interface where optical feedback is highest. Laser-induced explosion of ethanol droplets containing laser dye was observed at higher input intensities. Investigation of such laser-induced explosion was carried out using an optical multichannel analyzer (OMA) which can simultaneously provide spectral information from many points along a line. It has been determined that lasing is indeed confined within the droplet. Furthermore, it has been determined that the material containing flurorescent dye can be ejected from the droplet in the form of plumes which are located in front of the shadow face and/or in back of the illuminated face. These results have been reported in a special issue of the Academia Sinica (publication #3) and in the Proceedings of the Second International Laser Science Conference (publication #4).

2. Nonlinear Optical Spectroscopy of Droplets

Since all nonlinear optical interactions are nonlinearly dependent on the intensity, it was essential to make a detailed calculation of the

internal-field and near-field distribution for micrometer-size droplets when electromagnetic waves with submicrometer wavelengths are incident on the droplets. The main conclusion of this study is that the droplet acts as a lens that causes a high field at three locations: (1) just outside the droplet, in front of the shadow face with an intensity enhancement of 10^3; (2) just within the droplet, near the shadow face with an intensity enhancement of 10^2; and (3) just within the droplet, near the illuminated face with an intensity enhancement of one-third that of the conjugate spot within the shadow face. A detailed report of these calculations and experimental verification of the internal and external high-intensity spots will be published in Applied Optics (publication #5).

The SRS line shape of the O-H stretching mode of H_2O in water droplets containing structure-breaking and structure-forming ions was investigated. The SRS radiation from the anions (NO_3^- and ClO_4^- within the droplet) can be so intense that it can serve as the pump to excite SRS of H_2O and the next higher order Stokes of the anions. Reports of these observations were made at two international conferences: the First World Congress on Particle Technology (publication #6); and the Tenth International Conference on Raman Spectroscopy (publication #7). Another manifestation of the intense internal field strength is spectral broadening of the SRS and elastic scattering radiation. With CS_2 droplets, such linewidth broadening is due to phase modulation assoc iated with the intensity-dependent index of refraction ($n = n_o + n_2I$), which causes the liquid to have a time varying index of refraction during the time the Raman and input radiation is traveling around the

droplet circumference. A report of this result appeared in Optics Letters (publication #8).

It is generally accepted that the laser-induced breakdown of air or various gases is lowered by the presence of aerosols. However, controversy exists in the literature regarding the location at which the laser-induced breakdown is initiated, i.e., within the droplet or in the surrounding gas outside the droplet. Whereas previous work relied on taking photographs of the luminous flash associated with the breakdown in the droplet or in the gas, the present work made use of the OMA to provide spatially resolved spectra associated with the plasma continuum and the discrete atomic emission lines, e.g., hydrogen Balmer lines when the H_2O breaks down and singly ionized nitrogen N(II) lines when air breaks down. Depending on the breakdown strength of the liquid and the surrounding gas, breakdown can be initiated first in the gas and then in the liquid or breakdown can be initiated only within the liquid. The location of laser-induced breakdown has been determined by noting the location of the emission relative to the droplet interface. However, once breakdown in the liquid has occurred, the internal plasma absorbs the subsequent portion of the laser pluse, prevents the remaining laser radiation from reaching the gas in front of the shadow face, and produces an optical detonation wave which propagates toward the laser beam away from the illuminated face. Color photographs of the two plumes have been reproduced in an article highlighting our work at Yale [Science News 130, 408 (1986)]. Some of these results have also been published in Optics Letters (publication #9); others have been only recently submitted to the same journal (publication #10).

The internal and near-field calculations and the spatially resolved spectroscopy of the breakdown emission at the two plumes and within the droplet were helpful in providing information regarding the breakdown mechanism and the generation of plumes by the associated shock wave (propagating toward the front, away from the shadow face) and the optical detonation wave (propagating toward the laser, away from the illuminated face). The plasma density and temperature along a strip which encompasses the two plumes can be deduced from the emission line width (greatly Stark broadened) and relative emission ratios (assuming a local thermodynamic equilibrium). A preliminary report of these results has recently been submitted to Physical Review Letters (publication #11).

3. Three-Component Velocity Determination

Time-coded five-pulse laser volume holograms (in line) of silver-coated phenolic resin microballoons flowing in a turbulent field have been made. Time coding was used in order to distinguish the direction of the particle trajectory. Five pulses were used to ensure that the five bright dots were from the same particle flowing in a turbulent field. Application of a silver coating (by chemical deposition) on the microballoons significantly increased the contrast in the reconstructed hologram between the bright background and the particle. An automated optical pattern recognition scheme was used to transform the reconstructed volume hologram image into digital information in a computer and then to determine in an efficient manner the existence, location, and orientation of a straight-line segment formed by the five bright dots. After it has been determined that there is a straight line in the

x-y image (one plane in three dimensions) at a particular z location, the length and direction of the line provide values for v_x and v_y. Successive x-y planes are examined as a function of z. In principle, v_z can be determined from the five dots forming a straight line in successive x-y planes with different z values. Details of these results are currently being written up for submission to Applied Optics.

PUBLICATIONS

1. S.-X. Qian, J.B. Snow, H.-M. Tzeng, and R.K. Chang, "Lasing Droplets: Highlighting the Liquid-Air Interface by Laser Emission," Science 231, 486 (1986).

2. J.B. Snow, S.-X. Qian, and R.K. Chang, "Nonlinear Optics with a Micrometer-Size Droplet," Opt. News 12 (5), 5 (1986).

3. W.-F. Hsieh, H.-M. Tzeng, and R.K. Chang, "High Intensity Laser Interactions with Micrometer-Size Dye Droplets," in Special Issue of the Annual Report of the Institute of Physics, Academia Sinica (Taiwan), Vol. 16, 1986 in honor of Prof. Ta-You Wu's 80th birthday, p. 1.

4. R.K. Chang, "Micrometer-Size Droplets as Optical Cavities: Lasing and Other Nonlinear Effects," to be published in Proceedings of the 1986 International Laser Science Conference (American Institute of Physics, New York).

5. D.S. Benincasa, P.W. Barber, J.-Z. Zhang, W.-F. Hsieh, and R.K. Chang, "Spatial Distribution of the Internal and Near-Field Intensity of Large Cylindrical and Spherical Scatterers," Appl. Opt., in press.

6. J. Eickmans, S.-X. Qian, and R.K. Chang, "Detection of Water Droplet Size and Anion Species by Nonlinear Optical Scattering," in 1. World Congress on Particle Technology, Part I. Particle Characterization, K. Leschonski, ed. (NMA, Nuremberg, 1986), p. 125.

7. J.H. Eickmans and R.K. Chang, "Stimulated Raman Scattering Spectra from Single Water Droplets," in Proceedings of the Tenth International Conference on Raman Spectroscopy W.L. Peticolas and B. Hudson, eds. (University of Oregon, Eugene, 1986), p. 8-7.

8. S.-X. Qian and R.K. Chang, "Phase-Modulation-Broadened Line Shapes

from Micrometer-Size CS_2 Droplets," Opt. Lett. 11, 371 (1986).

9. J.H. Eickmans, W.-F. Hsieh, and R.K. Chang, "Laser-Induced Explosion of H_2O Droplets: Spatially Resolved Spectra," Opt. Lett. 12, 22 (1987).

10. W.-F. Hsieh, J.H. Eickmans, and R.K. Chang, "Internal and External Laser-Induced Avalanche Breakdown of Single Droplets in an Argon Atmosphere," submitted to Opt. Lett.

11. J.H. Eickmans, W.-F. Hsieh, and R.K. Chang, "Plasma Spectroscopy of H, Li, and Na in Plumes Resulting from Laser-Induced Droplet Explosion," submitted to Phys. Rev. Lett.

12. H.-M. Tzeng, M.B. Long, R.K. Chang, and P.W. Barber, "Size and Shape Variations of Liquid Droplets Deduced from Morphology-Dependent Resonances in Fluorescence Spectra," in Proceedings of the SPIE Particle Sizing and Spray Analysis Conference, Vol. 573 (SPIE, Bellingham, Washington, 1985), p. 80.

13. R.K. Chang, S.-X. Qian, and J. Eickmans, "Stimulated Raman Scattering, Phase Modulation, and Coherent Anti-Stokes Raman Scattering from Single Micrometer-Size Liquid Droplets," in Methods of Laser Spectroscopy, Y. Prior, A. Ben-Reuven, and M. Rosenbluh, eds. (Plenum Press, New York, 1986), p. 249.

14. S.X. Qian and R.K. Chang, "Multiorder Stokes Emission from Micrometer-Size Droplets," Phys. Rev. Lett. 56, 926 (1986).

15. J.B. Snow, J.-B. Zheng, and R.K. Chang, "Increased Sensitivity of a Vidicon Optical Multichannel Analyzer with a Detachable Electrostatic Image Intensifier," Appl. Opt. 25, 172 (1986).

PERSONNEL

Graduate Students

David Leach
Jian-Zhi Zhang

Postdoctoral Associates

Viroj Vilimpac, now working with Dr. Larry Goss at Systems Research Laboratories, Inc., Dayton, Ohio.

INTERACTIONS

"Nonlinear Optical Effects in Liquid Droplets: Results from High Intensity Lasers," ARO Workshop on the Interactions of Electromagnetic and Particle Beams with the Atmosphere, Las Cruces, NM, 1/28/86.

"Nonlinear Spectroscopy from Single Micrometer-Size Droplets," Sandia National Laboratory, Livermore, CA, 2/6/86.

"Nonlinear Optics of Microparticles," Korean Advanced Institute of Science and Technology, Seoul, Korea, 2/22/86.

"Light Scattering from Microdroplets," Korean Advanced Institute of Science and Technology, Seoul, Korea, 2/25/86.

"Laser Scattering from Micron-Size Particles," Workshop on Laser Spectroscopy and Techniques, National Central University, Chung-Li, Taiwan, 3/1/86.

"Nonlinear Interaction in Droplets," Academia Sinica, Nankang, Taiwan, 3/3/86.

"Stimulated Oscillations in Micron-Size Objects," National Tsing-Hua University, Hsinchu, Taiwan, 3/5/86.

"High Intensity Laser Interactions with Liquid Droplets," Chung-Shan Institute of Science and Technology, Lung-Tan, Taiwan, 3/7/86.

"Laser Scattering from Micron-Size Particles," Department of Physics, Chinese University of Hong Kong, Hong Kong, 4/3/86.

"Nonlinear Optical Interactions from Droplets," Fudan University, Shanghai, People's Republic of China, 4/11/86.

"Laser Interactions with Microparticles," Sichuan University, Cheng Du, People's Republic of China, 4/15/86.

"Nonlinear Optical Interactions in Liquid Droplets during High Intensity Laser Propagation," International Conference on Optical and Millimeter Wave Propagation and Scattering in the Atmosphere, Florence, Italy, 5/28/86 (invited talk).

"Droplet Characterization by Light Scattering," International Conference on Optical and Millimeter Wave Propagation and Scattering in the Atmosphere, Florence, Italy, 5/30/86 (invited talk).

"Investigation of Superradiant LDV Markers and Three-Component Velocity Mapping," Air Force Office of Scientific Research Workshop on Diagnostics of Reacting Flows, Stanford University, Stanford, CA, 6/17/86.

"Nonlinear Optical Effects and Breakdown of Water Droplets," CRDEC Scientific Conference on Obscuration and Aerosol Research, Edgewood Arsenal, MD, 6/25/86.

"Nonlinear Optical Scattering with Droplets," Sanders Associates, Nashua, NH, 7/18/86.

"Nonlinear Optical Spectroscopy with H_2O Droplets Containing NO_3^-," Conference on HAN-Based Liquid Propellant Structure and Properties, Aberdeen Proving Ground, MD, 7/29/86.

"Micrometer-Size Droplets as Optical Cavities: Lasing and Other Nonlinear Effects," Second International Laser Science Conference, Seattle, WA, 10/23/86 (plenary talk).

Discussion with Dr. Fred Quelle at ONR Office, Boston, MA, regarding the applicability of research to DoD, 7/28/86.

END

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